

# THz-field-induced transient magnetization and electric polarization in quantum paraelectric diamagnet $\text{KTaO}_3$

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Recently, Juraschek *et al.*<sup>1,2</sup> theoretically predicted that circularly polarized phonons should carry orbital magnetic moment even in diamagnetic or paramagnetic materials. The macroscopic orbital magnetic moment should be induced by the excitation of two perpendicularly polarized degenerate polar phonons using circularly polarized THz radiation. Unfortunately, experimental evidence of this phenomenon has been lacking.

We decided to pump the ferroelectric soft mode in quantum paraelectric  $\text{KTaO}_3$  single crystal using extremely intense pulses with frequency 0.7 THz. The pulses with a duration of about 10 ps were generated by the free-electron laser at the TELBE beamline in the Helmholtz Zentrum in Rossendorf. The transient magnetic moment on the picosecond time scale was measured via the transient birefringence at a probe wavelength of 800 nm, expected to induce the Faraday effect. A possible breaking of inversion symmetry was sensed by measuring the second harmonic generation at 400 nm. The experiments were performed between 10 and 300 K.

At low temperatures, the THz pump frequency was close to the soft-mode frequency, and a fairly strong transient birefringence was observed. However, THz pump-induced transient birefringence was detected not only using a circularly polarized THz beam, but also upon pumping with a linearly polarized beam. We explain this by the THz-field-induced Kerr effect which was recently observed also in the quantum paraelectric  $\text{SrTiO}_3$ .<sup>3</sup> In  $\text{KTaO}_3$ , a transient polarization and the related broken inversion symmetry were proved also by measuring the transient second harmonic generation.

The excited phonons were polarized in the sample plane, so the induced magnetization should be perpendicularly to the sample plane. In this case, theoretically, the magnitude of the Faraday signal should be the same after pumping using clockwise and anticlockwise circularly polarized THz radiation. Surprisingly, we detected a marked difference. We explain our observations by a combination of Faraday and Kerr effects, combined with the directional dichroism which is allowed in non-centrosymmetric diamagnetic systems in the presence of magnetic field.<sup>4</sup> In our case, the magnetic field was induced by the circularly polarized phonons.

In summary, we demonstrated that a transient polarization and magnetization can be induced simultaneously, by a strong excitation of polar phonons in a diamagnetic and quantum paraelectric crystal.

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<sup>1</sup> D. M. Juraschek, M. Fechner, A. V. Balatsky and N. A. Spaldin. *Phys. Rev. Mat.* **1**, 014401 (2017).

<sup>2</sup> D.M. Juraschek, N.A. Spaldin, Orbital magnetic moments of phonons *Phys. Rev. Mat.*, **3**, 064405 (2019)

<sup>3</sup> X. Li, T. Qiu, J. Zhang, E. Baldini, J. Lu, A. M. Rappe, and K. A. Nelson. Terahertz field-induced ferroelectricity in quantum paraelectric  $\text{SrTiO}_3$ . *Science* **364**, 1079-1082 (2019).

<sup>4</sup> A. I. Nikitchenko and R. V. Pisarev. Magnetic and antiferromagnetic nonreciprocity of light propagation in magnetoelectric  $\text{CuB}_2\text{O}_4$ . *Phys. Rev. B* **104** (2021).